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A One-Kilogram Quartz Resonator as a Mass Standard

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***Abstract*—The SI unit of mass, the kilogram, is defined by a single artifact, the International Prototype Kilogram. This artifact, the primary mass standard, suffers from long-term instabilities that are neither well understood nor easily monitored.**

A secondary mass standard consisting of a 1-kg quartz resonator in ultrahigh vacuum is proposed. The frequency stability of such a resonator is likely to be far higher than the mass stability of the primary mass standard. Moreover, the resonator would provide a link to the SI time-interval unit. When compared with a laboratory-grade atomic frequency standard or GPS time, the frequency of the resonator could be monitored, on a continuous basis, with 10^{-15} precision in only a few days of averaging. It could also be coordinated, worldwide, with other resonator mass standards without the need to transport the standards.

I. Introduction

"The unit of mass, the kilogram, is the last remaining SI base unit defined by an artifact" [1]; the International Prototype Kilogram (IPK) is a 90% platinum–10% iridium alloy cylinder. It is available only at the Bureau International des Poids et Mesures (BIPM), in Paris. Mass standards to be calibrated, including the national standards, must be taken to the BIPM for calibration.

The IPK, placed in service in 1889, suffers from long-term instabilities that are neither well understood nor easily monitored [2]. Since 1889, its mass and those of its official copies have drifted apart by as much as 70 μg; i.e., up to an average of 0.3 μg per year; or 3×10^{-10}/y. One difficulty in determining the instability is that measurements rely on the assumption that the mass artifacts are not changing together. Until a more stable standard is found, common-mode, or correlated, long-term drift cannot be detected. 1-kg quartz resonators could independently monitor the primary standard's stability.

II. Primary Standards and the Role of Frequency

Because accuracy and stability of frequency can be measured at levels below 10^{-15}, many times better than other units of measure, methods are currently being sought that transduce other units to frequency [3]. In metrology laboratories, instability is quantified as a Type A uncertainty.

Quartz resonators have been an integral part of high-precision metrology ever since the definition of the second as the duration of 9 192 631 770 periods of the radiation corresponding to the transition between the two hyperfine levels of the ground state of the caesium 133 atom. A little-recognized and underappreciated fact is that the output signal of a cesium (Cs) frequency standard is produced by a high-stability quartz oscillator. The quartz oscillator frequency, through a so-called multiplier chain, is controlled by a frequency-lock loop (FLL) that obtains the expected frequency of a cesium resonance [4]–[6].

As an example of units conversion, consider the Josephson Voltage Standard (JVS), which uses the Josephson-junction effect to linearly transduce a frequency to a voltage [7]. A JVS typically has a total normalized uncertainty, ΔV/V, lower than 10^{-10}. Much of this uncertainty is in the connection between the JVS on-chip array to the user. In any case, a JVS is three orders of magnitude better than the accuracy and stability of the best oven-stabilized Zener-diode reference. Another point is that Zener-based voltage standards require yearly calibrations versus the official US 10-V primary standard held at NIST or another JVS. In contrast, JVS accuracy and stability are directly the accuracy and stability of the frequency reference, which is far better than its 10^{-10} voltage uncertainty.

We propose creating a quartz resonator as a secondary mass standard. A vast quantity of literature on the topic of quartz resonators and oscillators exists. Much of it can be applied to the objective of a secondary mass standard. An accurate primary mass standard using transduction of frequency to mass of quartz is not feasible; however, the total mass departure of a quartz resonator after a calibration could be orders of magnitude smaller than that of other mass standard reference materials (SRMs) because the oscillator frequency can be continuously monitored and traced to international standards, e.g., by means of the Global Positioning System (GPS), to inaccuracy levels substantially lower than 10^{-10}/y.

III. Quartz Crystal Resonators

More than 10^9 resonators are produced annually for frequency control and timing applications. Some resonators are also produced for mass sensing by means of quartz crystal microbalances (QCMs) [8], [9]. For example, QCMs have been used for decades to monitor film thickness during film deposition.

In QCMs, the frequency shift, Δf, caused by a small change in resonator mass, Δm, can be expressed, to a

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good approximation, as $\Delta f/f = -\Delta m/m$. Therefore, by measuring Δf, one may readily calculate the mass Δm deposited onto, or removed from, a QCM. For example, a high-stability resonator made for some of the most demanding applications, such as for communication and navigation satellites, is a 5-MHz fifth-overtone SC-cut resonator. The frequency noise (Allan deviation) of such a resonator, at the optimum averaging time (at the noise floor), is below 1×10^{-12}. The thickness of such a resonator is 2 mm in the center (they are contoured) and its active volume weighs 0.2 g. A 1×10^{-12} frequency fluctuation is therefore equivalent to a mass fluctuation of 2×10^{-13} g. With tiny, nanoelectromechanical resonators, single-molecule adsorption events can be detected [10].

The expression relating Δm to Δf does not hold for nonuniform mass loading [11], such as a dust particle on the surface, however, because the resonator mass standard is intended to be kept in ultrahigh vacuum, nonuniform mass loading is unlikely to be a significant problem. Even if there is some nonuniform mass loading on the resonator surfaces initially, it is only changes in the mass loading, not the mass loading itself, that affect the resonator's frequency stability.

IV. One-Kilogram Resonator

Is making a 1-kg resonator feasible?[1] Although making such a resonator would not be easy, no new technology would be required. The main barrier is the availability of sufficiently large, single-crystal quartz of high quality. Although 10-kg cultured quartz crystals are available, they are not large enough because their aspect ratios limit the dimensions of the quartz plate needed for a 1-kg resonator. (The plate of the mass standard being proposed here must be cut along specific directions with respect to the crystallographic axes and should be seed-free.)

Cultured quartz crystals of high perfection are not normally grown large enough for making a 1-kg SC-cut resonator, but they could be. The technology for growing such a crystal is readily available. It would take a special growth run, and to grow crystals of the highest perfection would take about three years [13]. Using natural quartz would be a quicker and, possibly, a better solution. Using a rectangular plate instead of the conventional circular plate would also ease the size requirement.

Because the density of quartz is 2.65 g/cm^3, a 1-kg resonator would require 377 cm^3 of quartz. The largest and most stable resonators being made today are the 5-MHz fifth-overtone SC-cut resonators. These use contoured circular plates 15 mm in diameter × 2 mm thick (in the center). One way to make a 1-kg resonator would be to scale up the dimensions by about a factor of 10. One manifestation of this would result in a frequency of about 180 kHz [14].

Another way to ease the material availability problem is to, at least initially, use a 100-g resonator. A 100-g standard can be compared with a 1-kg standard with only a small loss of accuracy [15]–[17]. Mass comparators are commercially available [18].

V. Resonator Stability

A major difference between resonators made in the past and those that would be made for a mass standard is that the resonators made in the past had to be rugged enough to withstand the trials of everyday use, especially shock, vibration, and temperature changes. For example, the highest-stability resonators have been those that were made for space applications, where the resonators must withstand the severe environmental conditions present during rocket launch. The aging rate reported for one type of such resonators is 9×10^{-10} total normalized frequency change in 8 years, or 2×10^{-13}/day (8-year average) [19], and 8×10^{-14}/day [20] for another type. A linear extrapolation of the 8 years of aging to 100 years results in 1×10^{-7} per 100 years. Normally, resonators age as a logarithmic function of time [21], [22]. A logarithmic extrapolation would result in $<1 \times 10^{-7}$ per 100 years.

To put these numbers into perspective, the instability of the IPK has been reported [2] to be about 50 μg/kg/100 y = 5×10^{-8} per 100 years! (The change may be even larger if the prototype and its "identical" copies are all changing together.)

A 1-kg resonator made especially for mass metrology would exist in an ultrahigh vacuum, in a benign laboratory environment. It would not have to be ruggedized. This would permit designs that could not be used for everyday applications. Moreover, stability scales with size. The most stable resonators have also been the largest ones; i.e., those with the smallest surface-to-volume ratio. For example, the adsorption and desorption of surface contaminants is one instability mechanism the effect of which decreases when the surface-to-volume ratio is smaller.

In addition, the long-term stability (aging) of a resonator generally improves with time [21], [22]. Therefore, the 1-kg resonator can reasonably be expected to exhibit substantially better long-term, as well as short-term, stability than any resonator made in the past.

Reference [23] includes a review of the instability mechanisms of quartz resonators and oscillators, a discussion of the fundamental limits of each mechanism, and suggestions for approaching the limits.

After applying the best available design and fabrication techniques, the resulting resonator would probably exhibit a far higher stability than any previously made resonator,

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[1] Historical footnote: Prior to the selection of platinum–iridium alloy for the IPK, in 1872, the organization making the selection, the International Metre Commission, "examined three other possibilities, namely quartz, glass and other pure metals. One of the members of the Commission, Herr, was a partisan of quartz because of its ideal crystal structure that by its very nature, he said, should be stable" [12].

but, even if that turns out to be not the case, the current state-of-the-art resonator stabilities would still provide a stability that is comparable to that of the IPK and its "identical" copies. Ultimately, we want to determine the drift of $\Delta m/m$ with the best precision. In a lab-grade Cs standard, the frequency drift is at levels of 10^{-16}, for years.

The stability of an ensemble of resonators can exceed the stability of any resonator in the ensemble (provided that the instability mechanisms are uncorrelated) [24], [25]. Moreover, because resonators of different designs respond differently to the various instability mechanisms, by including in the ensemble resonators of different types, e.g., third-overtone and fifth-overtone SC-cut as well as AT-cut, GT-cut, and tuning fork resonators, one may gain insights into the instability mechanisms.

A critical shortcoming of the IPK is that it lacks two important properties of standards. IPK accuracy cannot be decentralized, in the sense that it is not "readily available to all, constant throughout time and space, and easy to realize" [3]. The IPK is not portable. In contrast, measurements of quartz oscillator frequencies can easily be made using an atomic oscillator or GPS time as a reference. GPS time is currently maintained to within 100 ns (10^{-7} s) of the primary standards that contribute to UTC. Thus, the frequency uncertainty per year would be 3.2×10^{-15} relative to the most accurate reference.

The frequency of a quartz resonator is sensitive to ambient pressure. (In fact, some of the most sensitive pressure sensors are based on quartz resonators [26].) The dependence of frequency on atmospheric pressure can be readily measured with high accuracy. The proposed resonator mass standard would operate in vacuum. The same is true of the two primary mass standards under development, the watt balance and Avogadro project. The establishment of traceability of the mass scale in air to the realization in vacuum is critically important. Methods of accomplishing that traceability are known, and improvements are being developed [1], [18], [27], [28]. Similarly, a method will have to be developed for comparing a quartz resonator's mass in vacuum to a mass standard in air.

VI. Worldwide Coordination of Mass Standards

There was a time when frequency standards had to be transported to calibrate a remotely located secondary standard. The use of GPS and other time synchronization techniques have eliminated the need to transport a high-accuracy frequency standard for remote calibrations.

Similarly, with quartz resonators as secondary mass standards, worldwide comparisons of mass standards could be performed with no need to transport the standards. The frequencies of the quartz oscillators containing the resonators could be compared on a continuous basis, automatically, with an accuracy no worse than 10^{-12} at sample times of a few thousand seconds.

VII. Conclusions

A 1-kg quartz resonator that is referenced to time synchronization services, such as GPS, or to an atomic frequency standard, has the potential to provide a secondary mass standard that is as stable as, if not more stable than the IPK and its official copies, but without the need for a physical calibration. Moreover, quartz-resonator frequency drift, and hence mass, can be measured continuously with remarkable precision. The frequency prediction error in the long term can have a precision as good as 10^{-16}, which is substantially better precision than the long-term predicted mass stability of IPK and its copies.

The resonator would provide a link to the SI time-interval unit. The frequency of the resonator could be monitored, on a continuous basis, with ultrahigh precision. It could also be easily compared with other resonator mass standards, automatically, with no need to transport mass standards.

The authors hope that the ideas presented here will stimulate further studies that may lead to fundamentally better mass calibrations.

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